

PATENT ABSTRACTS OF JAPAN

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(71)Applicant : TOSHIBA CORP

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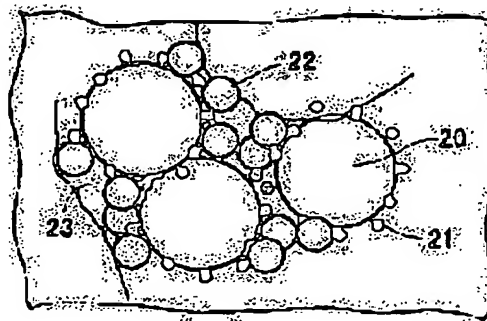
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(54) FUEL CELL AND FUEL CELL ELECTRODE MEMBER

(57)Abstract

PROBLEM TO BE SOLVED: To provide an electrode member for a high output and small size fuel cell that maintains a high stability for a long period and has a high proton conductivity.

SOLUTION: This is an electrode member which is equipped adjoining the electrolyte membrane of a fuel cell in which fuel is directly supplied. It comprises an electron conducting particle (20), a catalyst (21) and a proton conducting body, and the proton conducting body comprises an inorganic proton conducting solid particle (22) which forms a framework of the proton conducting pass from the surface of the electron conducting particle to the electrolyte membrane and an organic proton conducting body (23) which links between the inorganic proton conducting solid particles, and the catalyst and the inorganic proton conducting solid particle.



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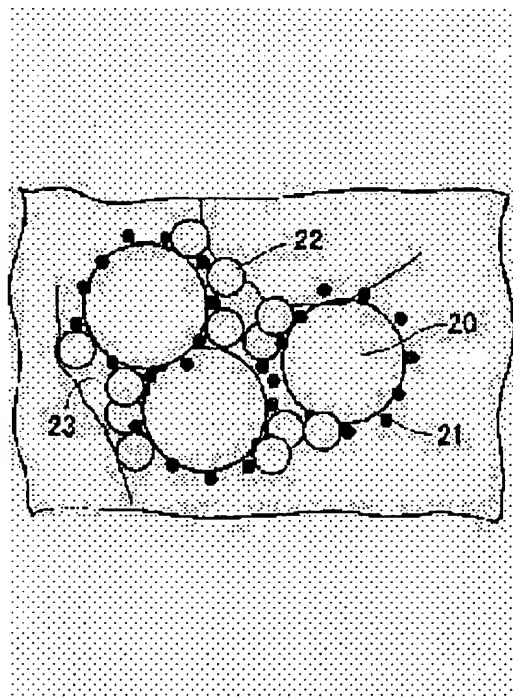
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CLAIMS

[Claim(s)]

[Claim 1] The anode electrode and cathode electrode which pinch an electrolyte membrane and said electrolyte membrane are provided. It is the fuel cell with which a fuel is introduced into said anode electrode. Said anode electrode and said cathode electrode Said proton conductor in either [at least] said anode electrode or said cathode electrode, including respectively an electronic conductor particle, a catalyst, and a proton conductor The inorganic proton conductor solid particulate which forms the frame of the proton electric conduction pass from the front face of said electronic conductor particle to said electrolyte membrane, The fuel cell characterized by including the organic proton conductor which connects between said inorganic proton conductor solid particulates and between said catalyst-inorganic proton conductor solid particulates.

[Claim 2] It is the electrode member characterized by to be the electrode member in which a fuel is prepared in contact with the electrolyte membrane of the fuel cell supplied directly, and to include the inorganic proton conductor solid particulate in which said proton conductor forms the frame of the proton electric conduction pass from the front face of said electronic conductor particle to said electrolyte membrane including an electronic conductor particle, a catalyst, and a proton conductor, and the organic proton conductor which connects between said inorganic proton conductor solid particulates and between said catalyst-inorganic proton conductor solid particulates.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the electrode member used for this in a fuel cell.

[0002]

[Description of the Prior Art] Since it is efficient as an independent power plant in recent years, the fuel cell attracts attention as a cell which was suitable in order to carry out long duration actuation of the various small devices. While to attain a miniaturization is desired, and such a fuel cell improves the polar zone for that purpose and raises an output, it is required to secure long term stability. In the polar zone of a fuel cell, the anode electrode and the cathode electrode are usually prepared so that an electrolyte membrane may be inserted. The anode electrode and the cathode electrode consist of a catalyst for reforming a fuel, and the electronic conductor and proton conductor for taking out the electron and proton which reforming was carried out and were generated. In order to raise effectiveness, in addition to raising catalyst ability - and the amount of catalysts per unit area, improvement in catalyst use effectiveness is also indispensable. In order to raise catalyst use effectiveness, the proton and electron which were generated on the catalyst must be taken out efficiently, and must be made to conduct current. That is, it becomes important further that a catalyst, proton electric conduction pass, and a catalyst and an electronic conductor have surely touched and that proton electric conduction pass spread in the shape of a network, and has connected from a catalyst to the electrolyte membrane.

[0003] The organic proton conductor is used as current and proton electric conduction pass. This organic proton conductor has not employed that property efficiently enough, although the proton conductivity of the ingredient itself is high. This is because it originates in condensation of an electronic conductor occurring in case an electronic conductor and a proton conductor are mixed and it is made to distribute, or a crack arising when drying an electrode and is hard coming to take contact on a catalyst and proton electric conduction pass. That is, in the electrode containing an organic proton conductor, since the network of the proton electric conduction pass from a catalyst to an electrolyte is not formed enough, original proton conductivity and also the catalyst ability of a catalyst are not fully utilizable, either. Therefore, amelioration of electrode structure or the manufacture approach is needed. Moreover, in the case of a fuel cell like DMFC (a methanol direct mold fuel cell, Direct Methanol Fuel Cell) which contains a methanol in a fuel especially, the problem that an organic proton conductor will deteriorate much more by the alcoholic component has occurred.

[0004] On the other hand, since an inorganic proton conductor is strong and can also desire reinforcement for fuel-proof nature, development is performed. However, the present inorganic proton conductor can be produced upwards only with a sol gel process, and proton conductivity is not enough and it is 1/10 or less engine performance of an organic conductor efficiently. Therefore, an inorganic proton conductor cannot be used for the fuel cell expected high power with the present condition. Furthermore, when producing an inorganic proton conductor with a sol gel process, contraction of an electrode arises in the case of gelation, there is a problem that the configuration as an electrode cannot be maintained, and electrode production with an inorganic proton conductor is difficult in the present

process.

[0005]

[Problem(s) to be Solved by the Invention] As mentioned above, formation of a up to [mixing, distribution, and a current collection member with a catalyst and an electronic conductor] is difficult for the organic proton electric conduction pass used with the anode electrode and the cathode electrode in the conventional fuel cell. This is because the crack which the wettability of an electronic conductor and an organic proton conductor cannot distribute easily bad, and produces in an electrode surface further at the time of desiccation is not avoided. Originating in this, the contact condition of a catalyst and a proton conductor is not enough, and the proton electric conduction pass with which it was formed has many parts divided between the current collection section and an electrolyte membrane. On the other hand, an inorganic proton conductor has low proton conductivity, and since the production process is not established, it cannot form the electrode of the configuration stabilized using this.

[0006] Then, this invention aims at offering high power and a small fuel cell while it maintains high stability over a long period of time.

[0007] Moreover, this invention is an electrode member for high power and a small fuel cell, and it aims at offering the electrode member which has high proton conductivity while it maintains high stability over a long period of time.

[0008]

[Means for Solving the Problem] In order to solve the above-mentioned technical problem, this invention possesses the anode electrode and cathode electrode which pinch an electrolyte membrane and said electrolyte membrane. It is the fuel cell with which a fuel is introduced into said anode electrode. Said anode electrode and said cathode electrode Said proton conductor in either [at least] said anode electrode or said cathode electrode, including respectively an electronic conductor particle, a catalyst, and a proton conductor The inorganic proton conductor solid particulate which forms the frame of the proton electric conduction pass from the front face of said electronic conductor particle to said electrolyte membrane, The fuel cell characterized by including the organic proton conductor which connects between said inorganic proton conductor solid particulates and between said catalyst-inorganic proton conductor solid particulates is offered.

[0009] This invention is an electrode member in which a fuel is prepared in contact with the electrolyte membrane of the fuel cell supplied directly, and contains an electronic conductor particle, a catalyst, and a proton conductor. Moreover, said proton conductor The inorganic proton conductor solid particulate which forms the frame of the proton electric conduction pass from the front face of said electronic conductor particle to said electrolyte membrane, The electrode member characterized by including the organic proton conductor which connects between said inorganic proton conductor solid particulates and between said catalyst-inorganic proton conductor solid particulates is offered.

[0010] In this invention, an electronic conductor particle and a catalyst need for it to be in contact or to surely have adhered among the electronic conductor particle mentioned above, a catalyst, and a proton conductor. Moreover, it is also required for the catalyst and the proton conductor to surely have touched or adhered.

[0011] Hereafter, this invention is explained to a detail with reference to a drawing.

[0012] The partial schematic diagram of an example of the fuel cell of this invention is shown in drawing 1 R> 1.

[0013] In drawing 1, the 1st and 2nd electrodes 11a and 11b are formed in both sides of an electrolyte membrane 10, and 1st current collection section 12a and 2nd current collection section 12b are further prepared on these electrodes, respectively. As for the 1st and 2nd electrodes 11a and 11b, in this invention, a proton conductor [in / including an electronic conductor particle, a catalyst, and a proton conductor / at least / one side of such an electrode] contains the inorganic proton conductor solid particulate which has the following functions, and an organic proton conductor, respectively. That is, an inorganic proton conductor solid particulate forms the frame of the proton electric conduction pass from the front face of an electronic conductor particle to an electrolyte membrane, and an organic proton conductor connects between inorganic proton conductor solid particulates, and catalysts and inorganic

proton conductor solid particulates.

[0014] A fuel is supplied to either of the electrodes 11a and 11b which pinch an electrolyte membrane 10 so that it may illustrate. Although especially the fuel used is not limited but the thing of the shape of a gas and a liquid can be used, when liquid fuel, such as a methanol, is used, especially the effectiveness of this invention is demonstrated.

[0015] Such an inorganic proton conductor solid particulate and an organic proton conductor are roughly shown in drawing 2 with an electronic conductor particle and a catalyst. The catalyst 21 has adhered to the front face of the electronic conductor particle 20, and the inorganic proton conductor solid particulate 22 forms the frame of proton electric conduction pass so that it may illustrate. In order to connect such inorganic system proton conductor solid particulate 22 comrades and a catalyst 21, and the inorganic proton conductor solid particulate 22, the organic proton conductor 23 is arranged. It can be said that the organic proton conductor 23 is constituted so that the inorganic proton conductor solid particulate 22 may be covered.

[0016] That is, in this invention, the proton electric conduction pass which spreads in the shape of a network was realized, and chemical property-proof was high, and the inorganic proton conductor solid particulate 22 which is easy to distribute in a slurry with electronic electric conduction pass was used as a frame, and since the pass by the organic proton conductor 23 was formed so that it might be connected, it became possible to prepare stable proton electric conduction pass.

[0017] In this invention, although it will not be limited as an electronic conductor particle 20 especially if electronic electric conduction can be performed, the thing with low cost which has high electronic conductivity, for example, a carbon particle etc., is desirable. When using a carbon particle, as for the mean particle diameter, it is desirable that it is the particle of the range of 5nm - 1000nm, and it is more desirable that it is the range which is 10nm - 100nm.

[0018] When supporting a catalyst especially on a carbon particle, it is required to be a particle with a large specific surface area, and since specific surface area will generally also become large if particle diameter is fine, it is not desirable to use the big particle exceeding 1000nm. On the other hand, since distribution will become difficult in case it mixes with organic and an inorganic proton conductor and is made a slurry if the particle diameter of an electronic conductor particle is too fine, it becomes difficult to produce a smooth electrode with few cracks. And contact to an electronic conductor particle and a proton conductor worsens, contact of carbon particles also worsens, and it may be hard coming to carry out both proton electric conduction and electronic electric conduction. It becomes remarkable when the particle which has the mean particle diameter of less than 5nm especially is used.

[0019] Moreover, the specific surface area of an electronic conductor particle is so desirable that it is large, it is desirable that it is specifically more than 10m²/g, and it is more desirable that it is more than 40m²/g. It is because this has few amounts of catalysts supported with the support which has the surface area below 10m²/g when supporting a catalyst, as mentioned above and it is difficult to raise effectiveness.

[0020] As catalyst matter 21 used in this invention, platinum metals, such as platinum, a rhodium, a ruthenium, iridium, palladium, and an osmium, and the alloy of those are desirable. As for such a catalyst, to be supported by the front face of an electronic conductor particle is more desirable. Contact to the carbon particle as an electronic conductor particle is not only raised, but since catalyst particle diameter is also small and the supported catalyst is producible, its activity as a catalyst improves.

[0021] In addition, even when not supported by the electronic conductor particle, the fine thing of the particle diameter of a catalyst is desirable, and, specifically, its range of 1nm - 10nm is desirable.

Activity is [a catalyst particle simple substance] too high for the case of less than 1nm, and there is a possibility that handling may become difficult. If 10nm is exceeded, in order that the surface area of a catalyst may decrease and reaction sites may decrease in number on the other hand, there is a possibility that activity may fall.

[0022] Moreover, high particles of proton conductivity, such as 2OSiO [which was produced using a sol gel process as an inorganic system proton conductor solid particulate 22 which constitutes a proton conductor in this invention] 2, or SiO₂-P5 system, are desirable. As for the particle diameter of such an

inorganic system proton conductor solid particulate, it is desirable that it is smaller than the electronic conductor particle mentioned above, and it is 5nm - 100nm preferably.

[0023] As shown in the expanded sectional view of drawing 3 R> 3, as for the inorganic system proton conductor solid particulate 22, it is more desirable to have pore 24. It is desired for pore 24 to have pore size as detailed as 1nm - about 10nm. In case it mixes with the organic proton conductor 23 which set to the inorganic proton conductor solid particulate 22 which has such detailed pore 24, and was dissolved or distributed to the organic solvent, a solution enters in pore 24, and when it dries, it is more desirable that the pass of the organic proton conductor 23 is formed in a hole. Since it has the work which raises the proton conductivity of the inorganic system proton conductor solid particulate 22, and the work holding the pass of the organic proton conductor 23 prepared in the hole, as for pore 24, it is desirable to have the pore size of the range mentioned above.

[0024] The specific surface area of the inorganic proton conductor solid particulate 23 is so desirable that it is large, it is desirable that it is more than at least 10m²/g, and it is more than 100m²/g more preferably. This is because it is thought that the electric conduction mechanism of an inorganic proton conductor conducts current through the OH radical which is sticking to one on the front face. Since a surface adsorption OH radical increases when the specific surface area of an inorganic proton conductor solid particulate is large, proton electric conduction pass also increases and conductivity also improves.

[0025] The resin of sulfonic acid types, such as perfluoro sulfonic acid dissolved or distributed to the organic solvent as an organic proton conductor 23 on the other hand and a styrene-divinylbenzene system, is desirable.

[0026] In a proton conductor, it is desirable to mix an inorganic proton conductor solid particulate and an organic proton conductor so that the volume of the inorganic system proton conductor solid particulate 22 may be set to 0.1-1000 to the volume (solid content after desiccation) 1 of the organic proton conductor 23. When the rate of an inorganic proton conductor solid particulate is less than 0.1, it cannot become difficult to form the frame of proton electric conduction pass by inorganic proton conductor solid particulates, and the effectiveness which uses both organic and inorganic system cannot fully be acquired. On the other hand, when the rate of an inorganic system proton conductor solid particulate exceeds 1000, there is a possibility that it may become difficult to connect inorganic proton conductor particles with an organic proton conductor, and the contact in a proton conductor or contact for a proton conductor and a catalyst may become a defect. Furthermore, if the rate of an inorganic proton conductor solid particulate becomes high, the proton conductivity within an electrode will fall.

[0027] Moreover, as for the thickness of the organic proton conductor 23 constituted so that the inorganic proton conductor solid particulate 22 might be covered, it is desirable that it is 50nm or less, and it is more desirable that it is 10nm or less. This is because the part will deteriorate alternatively and degradation of the engine performance by the crack of an electrode, fragmentation of proton electric conduction pass, etc. with the passage of time will occur, if an organic proton conductor condenses and exists. This degradation is remarkably produced, when liquid fuel is used.

[0028] In the electrode member of this invention, when the volume of the appearance of an electronic conductor particle is set to 100, as for the mixing ratio of an electronic conductor particle and organic-inorganic proton conductor, it is desirable for the range of the volume of the appearance of organic-inorganic proton conductor to be 0.01-1000. If the ratio of proton electric conduction pass becomes less than 0.01, a big output cannot be obtained in order for the current which it becomes difficult to fully form wrap pass and can take out the front face of an electronic conductor particle to fall. On the other hand, if the rate of organic-inorganic proton conductor exceeds 1000, resistance of the polar zone increases and an output cannot be taken too.

[0029] The electrode member in this invention can be prepared by mixing each component with a mixing ratio which was mentioned above, and the obtained electrode member is contacted or pasted up on an electrolyte membrane.

[0030] The inorganic electrolyte produced as an electrolyte membrane which an electrode member contacts or pastes up with sulfonic acid type ion exchange resin, such as perfluoro sulfonic acid and a styrene-divinylbenzene system, and a sol gel process is desirable. Furthermore, it is more desirable that

in DMFC processing is performed on the surface of an electrolyte membrane so that the crossover of a fuel may not occur. It is a proton conductor like PBI, and, specifically, it is possible to coat a front face with the ingredient which a fuel cannot penetrate easily.

[0031] In addition, in the polar zone containing an electrode member and an electrolyte membrane, the electrolyte membrane, the anode electrode, and the cathode electrode have touched, or it is desirable that it is the pasted-up structure. As for an electrolyte membrane and an electrode, it is desirable to paste up using a hotpress etc., putting temperature and a pressure and to reduce contact resistance. Moreover, in order to raise bond strength, the solution which dissolved the organic solvent and the proton conductor may be applied to the adhesion side of each part material.

[0032] Moreover, in the fuel cell using a liquid like DMFC, or an organic gas-like fuel, although the fuel cell and electrode member of this invention are effective in carrying out long duration maintenance of the cell output, they are not limited to the fuel cell especially using said organic fuel.

[0033]

[Embodiment of the Invention] Hereafter, an example is shown and this invention is further explained to a detail.

[0034] (Example 1) The electronic conductor particle with which the carbon particle of 50nm of mean diameters was made to support a platinum-ruthenium (1:1) (amount of support 25wt%), and the catalyst was supported was obtained. Moreover, 2OSiO₂-P5 particle as an inorganic proton conductor solid particulate was produced with the sol gel process. The particle diameter of 2OSiO₂-P5 obtained particle was [4nm and the specific surface area of 10nm and pore size] 830m²/g.

[0035] in this way, prepared 10g of alloy support carbon particles and 2g of 2OSiO₂-P5 particles -- 6g of water, and organic proton electric conduction material (the Aldrich make, Nafion 20wt% solution) -- with 4g, it kneaded and the paste was obtained. This paste is applied on the carbon-electrode base material of a conductive porous body, and carried out room temperature desiccation, and the electrode member of this invention was produced.

[0036] (Example 1 of a comparison) The electrode member of the example 1 of a comparison was produced by the same technique using the same ingredient as the above-mentioned example 1 except not using an inorganic proton conductor solid particulate. However, organic proton electric conduction material set to 10g, and water was set to 8g.

[0037] The Nafion electrolyte membrane was pinched with the commercial cathode electrode, using the electrode member obtained in an above-mentioned example 1 and the above-mentioned example 1 of a comparison as an anode electrode. Furthermore, it pasted up at 140 degrees C using the hotpress, it constructed in the single cel of a fuel cell, and the cel was produced, respectively. In addition, cel area is 2 10cm.

[0038] About each cel, the methanol water-solution fuel of the concentration of 2 mol/l was supplied to the anode side by 4 ml/min as liquid fuel, and to the cathode side, ordinary pressure air was performed by 100 ml/min, and it generated electricity at delivery and 80 degrees C.

[0039] About each liquid-liquid object fuel cell, the relation between a current and an output is shown in the graph of drawing 4 . Among the graph of drawing 4 , Curve a is a result about the liquid fuel cell of this invention which used the electrode member of an example 1, and Curve b is a result about the liquid fuel cell which used the electrode member of the example 1 of a comparison. In the liquid fuel cell (curve a) of this invention, although it is stabilized to 0.4 A/cm² and an output can be taken out, in the liquid fuel cell (curve b) of the example 1 of a comparison, an output declines rapidly with the increment in a current, and only two or less 0.2 A/cm can take a current. As for this, the catalyst use effectiveness on a catalyst support particle originates in the area to which it is because the thing of an example 1 is higher, and the catalyst on a carbon particle touches proton electric conduction pass having bigger structure than the example 1 of a comparison. Moreover, in the liquid fuel cell of this invention, since the proton electric conduction pass used effectively is also increasing, when a current is taken, it turns out that the phenomenon in which an electrical potential difference falls is also suppressed.

[0040] Moreover, when the repeat trial was performed, in the case of the example 1 of a comparison, the engine performance fell for every count, but in the case of the example 1 using the electrode member

containing an inorganic proton conductor solid particulate and an organic proton conductor, the result of being almost comparable as the early engine performance was obtained. This originates in using the inorganic substance for proton electric conduction pass, and the resistance over the methanol fuel of an inorganic substance is because it is sharply high compared with macromolecules, such as Nafion.

[0041] Furthermore, as it was the following, it investigated about the performance degradation of an electrode.

[0042] First, the electrode member obtained in the example 1 and the example 1 of a comparison was left in the temperature of 60 degrees C, and air, and the accelerated test of performance degradation was performed. The neglect for one month in this condition is equivalent to the neglect for four months in the inside of room temperature air. After predetermined time passed, each electrode member was taken out, it used as an anode electrode and a cathode electrode, and the electrolyte membrane was pinched by these. Furthermore, it pressed, the polar zone was produced and cell measurement was performed.

[0043] The relation between the neglect time amount of an electrode and an output is shown in the graph of drawing 5. Among the graph of drawing 5, Curve c is a result at the time of using the electrode member of an example 1, and Curve d is a result at the time of using the electrode member of the example 1 of a comparison.

[0044] The early engine performance of the electrode (curve d) of the example 1 of a comparison is as small as 50 mW/cm², and, moreover, this engine performance has deteriorated with time. Even if degradation of the engine performance was saturated with the place which passed for three months and 12 months had passed it, the change beyond it was not checked.

[0045] On the other hand, in the case of an example 1 (curve c), the early engine performance is as large as 200 mW/cm², and the big change to an accelerated test 12 months after is not seen, but it turns out that degradation with the passage of time has not taken place. In the electrode member of this invention, since the inorganic proton conductor solid particulate and the organic proton conductor are included as shown in drawing 2 R> 2, it is because weatherability is improving.

[0046]

[Effect of the Invention] As explained in full detail above, while maintaining high stability over a long period of time according to this invention, high power and a small fuel cell are offered. Moreover, according to this invention, it is an electrode member for high power and a small fuel cell, and while maintaining high stability over a long period of time, the electrode member which has high proton conductivity is offered.

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DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

[Drawing 1] The fragmentary sectional view showing the configuration of a part of fuel cell of this invention.

[Drawing 2] The schematic diagram showing the structure in the electrode member of this invention.

[Drawing 3] The expanded sectional view showing the structure of the inorganic system proton conductor solid particulate in the electrode member of this invention.

[Drawing 4] The graphical representation showing the relation of the current of a fuel cell and electrical potential difference in connection with the example 1 and the example 1 of a comparison of this invention.

[Drawing 5] The graphical representation showing aging of the output of the fuel cell in connection with the example 1 and the example 1 of a comparison of this invention.

[Description of Notations]

10 -- Electrolyte membrane

11a, 11b -- Electrode

12a, 12b -- Current collection section

20 -- Electronic conductor particle

21 -- Catalyst

22 -- Inorganic proton conductor solid particulate

23 -- Organic proton conductor

24 -- Pore

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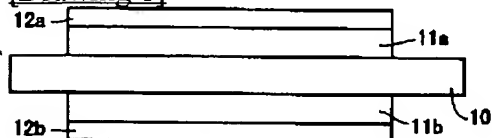
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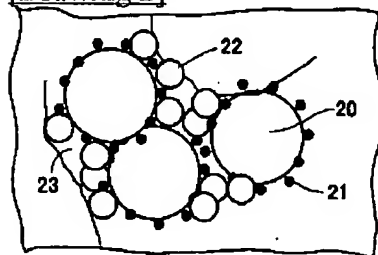
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DRAWINGS

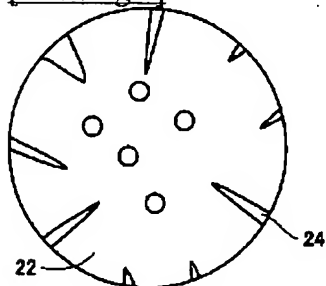
[Drawing 1]



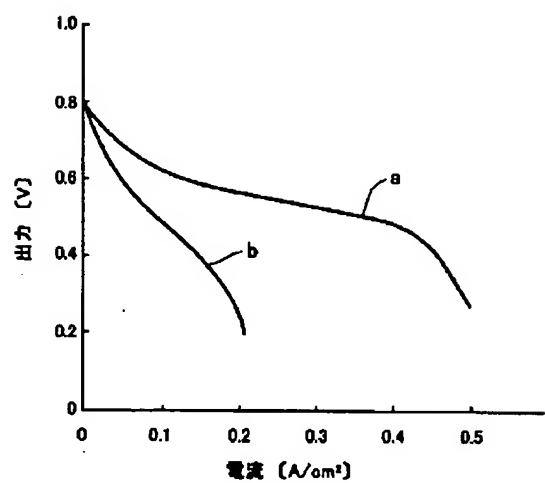
[Drawing 2]



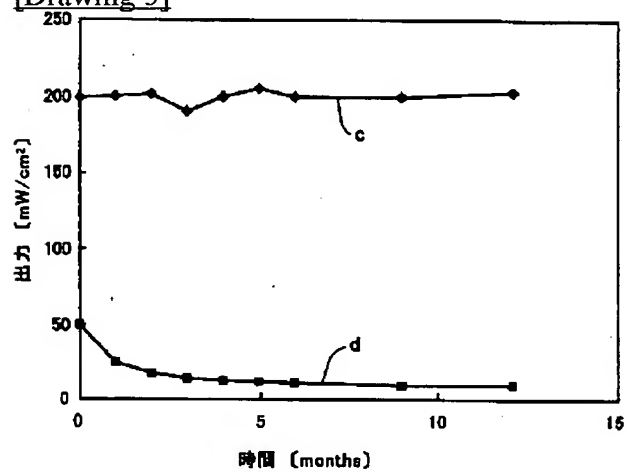
[Drawing 3]



[Drawing 4]



[Drawing 5]



[Translation done.]